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# A Performance Comparison of Individual and Combined Treatment Modules for Water Recycling

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## Abstract

An Advanced Water Recycling Demonstration Plant (AWRDP) was commissioned and constructed by the Queensland State Government in Australia. The AWRDP was used to study the effectiveness of a variety treatment processes in the upgrading of municipal wastewater for water recycling applications. The AWRDP consists of eight modules, each housing an individual specific treatment process. These processes are flocculation, filtration, dissolved air flotation, ozonation, biological activated carbon adsorption, microfiltration, reverse osmosis and ultraviolet disinfection. The individual performances of the treatment processes were determined, as well as their interdependence in series. A range of chemical water quality parameters were investigated. The study provides a broad process comparison on the basis of an important catalogue of these key parameters. This will be valuable in the selection and optimisation of treatment processes trains in full-scale water recycling applications.

Keywords: water recycling/reuse/reclamation, demonstration plant, membrane filtration, advanced oxidation, water quality.

## 1. Introduction

The Queensland Water Recycling Strategy (QWRS) is an initiative of the Queensland State Government (Gibson and Apostolidis, 2001). It provides a framework to encourage the adoption of sustainable water recycling to better manage water resources, and to support economic growth while protecting the environment and safeguarding public health. Among the objectives is the establishment of water recycling demonstration projects. An urgent need was identified for the community to see demonstrations of advanced technologies to treat recycled water to a quality suitable for a range of reuse applications. Accordingly, as a component of the QWRS, an Advanced Water Recycling Demonstration Plant (AWRDP) was commissioned and built (Figure 1). The AWRDP consists of eight modules each housing a different water treatment process. Each module

is designed as a relocatable unit so that the plant configuration may be adjusted and so that the entire plant may be easily transported to various sites throughout Queensland.

The primary objective of the AWRDP is to provide the community with a practical demonstration of water recycling technologies. This is aimed at encouraging community, industry and government confidence in the use of water treatment technologies and water recycling possibilities. A further aim is to provide opportunities for research to test the veracity and capabilities of the various treatment technologies. These objectives directly address some of the major current impediments to increased applications of municipal wastewater recycling in Australia (Khan *et al.*, 2004a).

This current research was initiated to investigate the water quality that can be achieved in various process configurations. This was undertaken in terms of effectiveness of treatment of selected physical and chemical parameters.

Most recycled water in Queensland comes from treated municipal sewage effluent. The principal uses of this water include irrigation of golf courses, public parks and gardens, playing fields, race courses, silviculture, viticulture, road construction, dust control, improved pastures, and crops such as sugar cane. Some additional applications recently being considered for the use of recycled water include power stations, oil refineries, ethanol production and irrigation of fresh farm produce. Accordingly, the monitoring program has focused on measuring those substances that relate to irrigation and potential industrial applications.

An area of rapidly growing concern for regulators is that of specific trace chemical contaminants. However, most current wastewater discharge and reuse guidelines are limited to bulk parameters such chemical oxygen demand (COD), biochemical oxygen demand (BOD), pH and total suspended solids (TSS). In many situations these simple parameters provide suitable surrogate indications of the likely presence of specific chemical species of concern. However, for more highly treated reuse waters, they may be of limited sensitivity. Furthermore, for some applications, an accurate assurance of specific chemical concentrations will be important.

National Guidelines for water reuse in Australia are currently under development (Cunliffe *et al.*, 2005). These are anticipated to be detailed in their regulation of specific chemical species. However, limits to bulk parameters are expected to maintain a key role as surrogate indicators of effective treatment. These guidelines will adopt the innovative risk management framework approach pioneered in the 2004 revision of the Australian Drinking Water Guidelines.

## 2. Materials and methods

### 2.1. The Advanced Water Recycling Demonstration Plant (AWRDP)

The Advanced Water Recycling Demonstration Plant consists of eight discrete treatment modules arranged in series with an additional storage module and control module. These may be operated in variable configurations:

1. Main plant feed tank with lime addition, flocculation, clarification and pH-adjustment by CO<sub>2</sub> addition
2. Dissolved air flotation (DAF)
3. Dual media filtration (DMF) consisting of alternating sand and anthracite layers.
4. Ozonation (O<sub>3</sub>). An OZAT CFS-2A ozone generator from Ozonia/Switzerland was used in the plant. The generator had a capacity of 80 g O<sub>3</sub>/h and requires 2 m<sup>3</sup> air/h air input. The dosage is 17.5 mg O<sub>3</sub>/L and the contact time 15 min.
5. Biological activated carbon filtration (BAC). The filtration unit consists of granular activated carbon in a fixed bed, colonised by micro-organisms growing in an attached bio-film.
6. Microfiltration (MF). A continuous microfiltration (CMF) unit supplied by Memcor consists of six capillary hollow fibre modules which are operated in dead-end mode. The membranes are operated in alternating filtration and backwash cycles and have an approximate cut-off of 0.2 µm. Back-flushing of the system is applied periodically and is dependent on the evolution of transmembrane pressure.

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7. Reverse osmosis (RO). Consists of two 8-inch pressure vessels carrying spiral wound modules with Trisep X-20<sup>®</sup> reverse osmosis membranes. Two 4-inch modules with different membranes were connected in parallel at both the inlet and the outlet of the 8-inch module for research purposes.
8. Ultraviolet disinfection (UV)

The feed water to the AWRDP is taken from Brendale Wastewater Treatment Plant (Pine Rivers Shire Council), which provides chlorinated municipal effluent treated by biological nutrient removal activated sludge process and sand filtration. Presently much of this municipal effluent is used to irrigate local sports fields. Recycled water from the AWRDP could potentially be used for a wide variety of applications requiring highly treated water. However, the plant's small scale and its location in a sports complex (for community visibility) during this investigation were not amenable for any such applications. Accordingly, the final AWRDP effluent was used to supplement the sports-field irrigation scheme at that time. The excess water, along with wastes and rejects from all units is currently discharged into the sewer system. The AWRDP is designed for an average throughput of 3.6-5.4 m<sup>3</sup>/h. A schematic drawing of the individual unit operations is given in Figure 2. Chlorine disinfection can be added to the process train, but was not included during this investigation due to the lack of any extensive distribution system and lack of requirement for the irrigation scheme. Issues such as skin contact of recreational users were discussed at the time, but mostly in the context of trace contaminants.

## 2.2. Sampling and analytical methods

Samples were taken from the feed of the AWRDP and the product streams of the various treatment options employed during the different phases and analysed in terms of the key parameters described below. Most of the analyses were undertaken following the methods outlined in "Standard Methods for the Examination of Water and Wastewater" (APHA, 1998). Total organic carbon (TOC) was determined using a Elementar High TOC II Carbon Analyser. The TOC was calculated by subtracting inorganic carbon (IC) from total carbon (TC). Both TC and IC were analysed by the thermal catalytic conversion of total carbon and carbonates (respectively) to carbon dioxide which was detected and quantified by non-dispersive infra-red (NDIR) spectroscopy. chemical oxygen demand (COD) was analysed by the closed reflux titrimetric method (APHA 5220(C)). Total suspended solids (TSS) was analysed gravimetrically (APHA 2540(D)). Turbidity was analysed by the Nephelometric method (APHA 2130(B)). Total dissolved solids (TDS) was analysed gravimetrically (APHA 2540(C)). Conductivity was analysed by an electrometric method (APHA 2510(B)). Nitrate was analysed by ion chromatography using a method developed by Queensland Health and Scientific Services (QHSS method QIS 18204). Trace metals (iron, aluminium, boron) were analysed by inductively coupled plasma atomic emission spectroscopy (QHSS method QIS 18197). Trace anions (bromide and iodide) were analysed by x-ray fluorescence spectrometry (QHSS method QIS 18235). Selected pharmaceutically active compounds (ibuprofen, gemfibrozil, naproxen, ketoprofen, metronidazole and carbamazepine) and estrogenic hormones (estrone, estradiol, ethinylestradiol) were analysed by gas chromatography-mass spectrometry with the full analytical details previously described (Khan *et al.*, 2004b).

## 2.3. Plant configurations

Five different plant configurations comprising key unit operations were tested within the project. These configurations are summarised in Figure 3.

Configuration 1 was regarded as a core concept, comprising biological activated carbon adsorption for organics removal, microfiltration membranes for particle and bacterial retention, followed by final UV disinfection.

Configuration 2 incorporated the ozonation module toward the front of the treatment stream of Configuration 1. In this configuration, ozonation was employed primarily as a pre-treatment step aimed at oxidising large molecule organics and thus enhancing the biological activated carbon adsorption performance.

Configuration 3 consisted of more conventional physico-chemical treatment options, comprising lime clarification, dissolved air filtration and dual media filtration followed by final UV disinfection. In this configuration, the initial treatment units primarily remove particulate and colloidal matter. Such a configuration is frequently employed in drinking water production and hence is a classical water treatment scheme.

Configuration 4 introduced reverse osmosis membranes in the water recycling scheme. Microfiltration removes residual particles after BAC and poses an excellent pre-treatment for the reverse osmosis modules. As microfiltration modules can be prone to blocking due to narrow internal channels, effective earlier treatment is essential. UV disinfection was again used as final stage to utilise a multi-barrier concept for pathogens. This type of water recycling scheme may be applied to produce high quality process water for irrigation in sensitive areas or industrial purposes.

Configuration 5 demonstrates the full capacity of the more advanced treatment options. It was employed to explore the limits of state-of-the-art technology with respect to water recycling. Ozonation was again applied as a pre-treatment to BAC, but was also anticipated to reduce the microbiological load prior to the membrane processes in order to minimise membrane bio-fouling. Activated carbon adsorption was intended to prevent membrane damage by residual ozone, as well as to adsorb or degrade ozonation by-products. The cost of this process train would be considerable, and the configuration may not be economically viable in many situations. However, the configuration was anticipated to provide high quality product with long term treatment stability and minimal chemical cleaning of the membranes.

## 3. Results and discussion

Four main categories of parameters were investigated in this study, namely 1. bulk organic parameters (TOC and COD); 2. inorganic parameters (nitrate, boron, aluminium, iron, bromide and iodide); 3. physical parameters (turbidity, suspended solids, total dissolved solids and conductivity) and 4. trace organic parameters (pharmaceutically active compounds and estrogenic hormones). These parameters were selected primarily to provide a variety of likely susceptibilities to the various treatment processes. For example degradative and oxidative processes could be assessed by their effect on parameters such as TOC and COD, while physical barriers provided by membranes would be most thoroughly tested by their effects on small cations and anions.

### 3.1. Bulk Organic Parameters

Each of these treatment process of Configuration 1 (Figure 4) contributed to some sequential reduction of the organic indicator parameters. TOC concentrations largely stabilised after BAC treatment. However, MF and UV further decreased COD, due to the retention of particulate matter and oxidation of organic components respectively.

A much sharper reduction in COD during BAC was observed in Configuration 2 (Figure 5) compared to Configuration 1 which did not include ozone pre-oxidation. This can be attributed to the enhanced adsorption and bio-degradation occurring in BAC after ozonation (Graham, 1999). During ozonation, unsaturated organic matter including aromatics are converted to smaller oxygenated molecules including aldehydes and carboxylic acids (Khan *et al.*, 2003). The smaller, oxygenated products of ozone treatment are significantly more biologically assimilable than their larger predecessors. In the presence of bromide ions, the formation of bromate can be a significant ozonation byproducts (von Gunten, 2003) which will require removal by subsequent treatment processes.

The conventional treatment sequence of Configuration 3 (Figure 6) resulted in a comparatively poor removal of the measured organic parameters. COD was only removed down to 25 mg/L compared to Configuration 1 (5.7 mg/L) and Configuration 2 (4.3 mg/L). While such treatment sequences may remove particulate and colloidal matter to a certain extent, they do not represent a physical barrier or chemical removal capacity for dissolved organic matter.

In Configuration 4 (Figure 7), BAC and MF provided similar organic parameters as Configuration 1. However, the RO membranes were observed to achieve significant further TOC and COD removal, in terms of both concentration and removal rate. The treatment sequence

achieved a removal of COD down to 1.67 mg/L. This result exceeds that achieved with ozonation, used in Configuration 2. However, some COD and TOC was observed remaining in the RO permeate, indicating that even RO processes have a limited retention for some organic components. del Pino and Durham (1999) described COD removal rates of about 80% for reverse osmosis membranes in water recycling applications, which is comparable to the results of this study for COD (72%) and TOC (73%). The production of a waste brine stream is a major hurdle in the implementation of pressure driven membrane processes, and requires adequate solutions for its treatment, reuse or discharge (Van der Bruggen *et al.*, 2003).

In Configuration 5 (Figure 8), the most comprehensive plant set-up was used with ozonation in addition to Configuration 4. The sharp COD drop after combined ozonation-BAC as was observed, as described for Configuration 2. Furthermore, the additional effect of the RO membrane is clear. This treatment scheme achieved a final COD concentration of 1mg/L and an overall removal efficiency of 96.7%.

The overall removal efficiency  $R_{total}$  of a particular process configuration, consisting of  $k$  unit operations with individual efficiencies  $R_i$ , was modelled by COD removal and calculated according to the following formula:

$$R_{total} = 1 - \sum_{i=1}^k (1 - R_i) \quad \text{with} \quad R_i = 1 - \frac{COD_{Product,i}}{COD_{Feed,i}} \quad (\text{Equation 1})$$

In Table 1 mean values for the COD-removal efficiencies  $R_i$  of the various unit operations are given. The interdependence between some unit operations can be clearly observed. As described, BAC performed more effectively following pre-ozonation. MF/RO and can be largely considered as a single functional unit since MF provides the essential pre-treatment for the RO module to protect the RO membranes from excessive fouling due to particulates and microorganisms. The effectiveness of MF/RO is reasonably independent of preceding modules. Most obvious is the dependence of UV on an efficient pre-treatment, while being most effective as a polishing stage after any BAC/membrane combination. A combination of UV with low-performance units like dual media filtration upstream is not effective, as was observed in Configuration 2. This observation accords with previous investigations of UV applications in water recycling (Lazarova *et al.*, 1999; Sakamoto *et al.*, 2001). Light scattering on particles and absorbance by organic matter are main reasons for such a dramatic loss in UV-facilitated oxidation of organics. The observed COD removal during the ozonation process was in the same range as that reported by Xu *et al.* (2002).

### 3.2. Inorganic and Physical Parameters

All of the applied unit operations were sufficient to remove turbidity and suspended solids to below the quantification limits, which were 1 NTU and 10 mg/L TSS, respectively.

Salinity parameters expressed as total dissolved solids (TDS) and conductivity were only significantly affected by the RO process. The feed conductivity was stable between 600 and 660  $\mu\text{S}/\text{cm}$  at 25°C, corresponding to approximately 350-380 mg/L TDS. The RO process reduced conductivity down to 15-27  $\mu\text{S}/\text{cm}$  and TDS to 8-11 mg/L. The corresponding retention for TDS is 97.5%, which is consistent with the specification for salt retention provided by the membrane supplier (98.5%). While this removal is significant, repeated application of low concentrations of non-degradable salts in effluents may risk a gradual build-up of soil salinity.

Feed concentrations of nitrate varied between 18 mg/L and 37 mg/L, significant denitrification in the biological activated carbon stage was observed on only one occasion. In this case the COD/Nitrate ratio of 1.8 was significantly higher than in all other cases where ratios between 0.5 and 1.3 were observed. RO operations achieved an average nitrate removal of 91% with final permeate concentrations of 0.6-2 mg/L. 81% nitrate removal has previously been reported during reverse osmosis (del Pino and Durham, 1999). Nitrogen is commonly a limiting nutrient for eutrophication in natural water ways. Therefore careful control of its release in forms including nitrate is important to prevent excessive algal growth.

The removal of the trace metals (boron, aluminium, iron) and anions (bromide and iodide) was investigated for the unit operations applied in Configuration 4. Iron, iodide and bromide displayed a

high retention by reverse osmosis membranes (71%-90%). However boron was not significantly retained due to its smaller atomic radius, influencing mobility in the membrane polymer. Boron was measured on the permeate side with concentrations of around 75  $\mu\text{g}/\text{L}$  in RO influent and 50  $\mu\text{g}/\text{L}$  in the RO effluent. This is an element that is known to accumulate in soils, but to date has not been given adequate attention in water reuse applications.

### 3.3. Trace Organic Contaminants

A range of pharmaceutically active compounds and estrogenic hormones were selected as representative anthropogenic organic compounds of emerging interest. Due to the more detailed analytical requirements, analysis of the selected trace organics was undertaken for Configuration 4 and Configuration 5 only.

In Configuration 4, the ozone treatment module was offline and samples were collected from the effluents of DMF, BAC, MF and RO/NF modules. Salicylic acid was identified in the effluents of every tested module with DMF (0.33  $\mu\text{g}/\text{L}$ ), BAC (0.23  $\mu\text{g}/\text{L}$ ), MF (0.13  $\mu\text{g}/\text{L}$ ), RO permeate (0.16  $\mu\text{g}/\text{L}$ ), and NF permeate (0.17  $\mu\text{g}/\text{L}$ ), as well as a concentrated solution in the RO retentate (2.21  $\mu\text{g}/\text{L}$ ). Gemfibrozil (0.18  $\mu\text{g}/\text{L}$ ) was measured in the DMF effluent, however none of the other analytes were measurable in the DMF, BAC, MF, RO permeate or NF permeate samples. On the other hand, most were concentrated enough to be measurable in the RO retentate with ibuprofen (0.31  $\mu\text{g}/\text{L}$ ), gemfibrozil (0.86  $\mu\text{g}/\text{L}$ ), naproxen (0.70  $\mu\text{g}/\text{L}$ ), ketoprofen (0.37  $\mu\text{g}/\text{L}$ ), estrone (0.092  $\mu\text{g}/\text{L}$ ), estradiol (0.0047  $\mu\text{g}/\text{L}$ ) and ethinylestradiol (0.0058  $\mu\text{g}/\text{L}$ ). Metronidazole and carbamazepine were not detectable in any of the samples.

Once the ozone module was brought online (Configuration 5), further testing was undertaken on the effluents of the DMF module (immediately before ozone), the  $\text{O}_3$  module and the BAC module (immediately after ozone). In these tests, salicylic acid was observed in DMF (0.65  $\mu\text{g}/\text{L}$ ),  $\text{O}_3$  (0.26  $\mu\text{g}/\text{L}$ ) and BAC (0.34  $\mu\text{g}/\text{L}$ ) effluents. All other analytes were below the limits of detection.

## 4. Conclusions

The AWRDP proved to be a successful tool in providing performance data for different unit operations and process trains. Interdependencies between the performance of different unit operations was clearly observed. For example, the influence of ozonation on subsequent biological activated carbon adsorption and the necessity of proper pre-treatment for an efficient application of UV-disinfection.

The combination of microfiltration and reverse osmosis proved to be a reliable upgrading stage with outstanding removal performance for the whole range of parameters considered. This process combination was completed by biological activated carbon adsorption and UV-disinfection to establish a core treatment sequence. While BAC provides a sink for adsorbable organic components, UV disinfection ensures hygienic quality.

A multi-barrier approach appears to be appropriate for the optimisation of overall treatment quality. At least three operational units appear optimum:

- (Biological) activated carbon adsorption
- Microfiltration
- UV-disinfection

This process configuration promotes organics removal but is not effective for a wide range of dissolved inorganic components which may only be removed with dense membranes like RO. Where a reduction in salinity and trace metal concentration is required reverse osmosis, or in some cases nanofiltration, will need to be applied. Such additions also offer additional safety in respect to virus removal and the elimination of polar micropollutants.

Ozonation was found to be a complimentary treatment stage but may not always be essential when the whole set of core treatment stages is applied. The conventional treatment stages like lime coagulation, dissolved air flotation and dual media filtration should only be considered as pre-treatment stages for BAC or ozonation and might prove useful to increase the efficiency and long-

term stability of downstream processes. Ozonation cannot be regarded as sufficient stand-alone treatment for the current scheme as most parameters were not sufficiently removed.

The selection of process configurations will be heavily influenced by economic considerations. The identified ‘core process’ of BAC/MF/UV will provide a high degree of safety in terms of chemical parameters with reasonable economics. While no economic studies were performed for this small scale demonstration plant, the investment cost for a 10,000 m³/d granular activated carbon plant have recently been quoted at around US\$ 286 per m³/d capacity with operation and maintenance costs estimated to be 0.14 US\$/m³ (AQUAREC, 2004). Ternes *et al.* (2003) have given cost estimates for large scale ozonation plants for effluent treatment as low as 0.04 €/m³ (investment + 10g/m³ ozone). Capital investment costs for full-scale MF systems as pre-treatment for RO units in wastewater reclamation have been quoted as US\$ 140-215 per m³/day for a 10,000 m³/day plant with operational costs about US\$ 0.10 per m³ (del Pino and Durham, 1999; AQUAREC, 2004).

Ultimately, process scheme selection in full scale treatment applications will be based on relevant guideline requirements in any particular case and risk assessment for potential exposure pathways including the use of the final product effluent. Water recycling guidelines are currently being developed world-wide.

### Acknowledgements

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### Tables and figures

Table 1: Mean Chemical Oxygen Demand (COD) removal efficiencies of unit operations (R<sub>i</sub> from Equation 1).

Unit operation	COD-Removal efficiency (%)
Lime coagulation	6
Dissolved air flotation	7
Dual media filtration	5
Ozonation	17
Biological activated carbon adsorption (BAC)	34
BAC with prior ozonation	61
Microfiltration (MF)	28
Reverse Osmosis	76
Ultraviolet disinfection (UV)	47
UV without prior BAC/MF	3



Figure 1: View of the Advanced Water Recycling Demonstration Plant

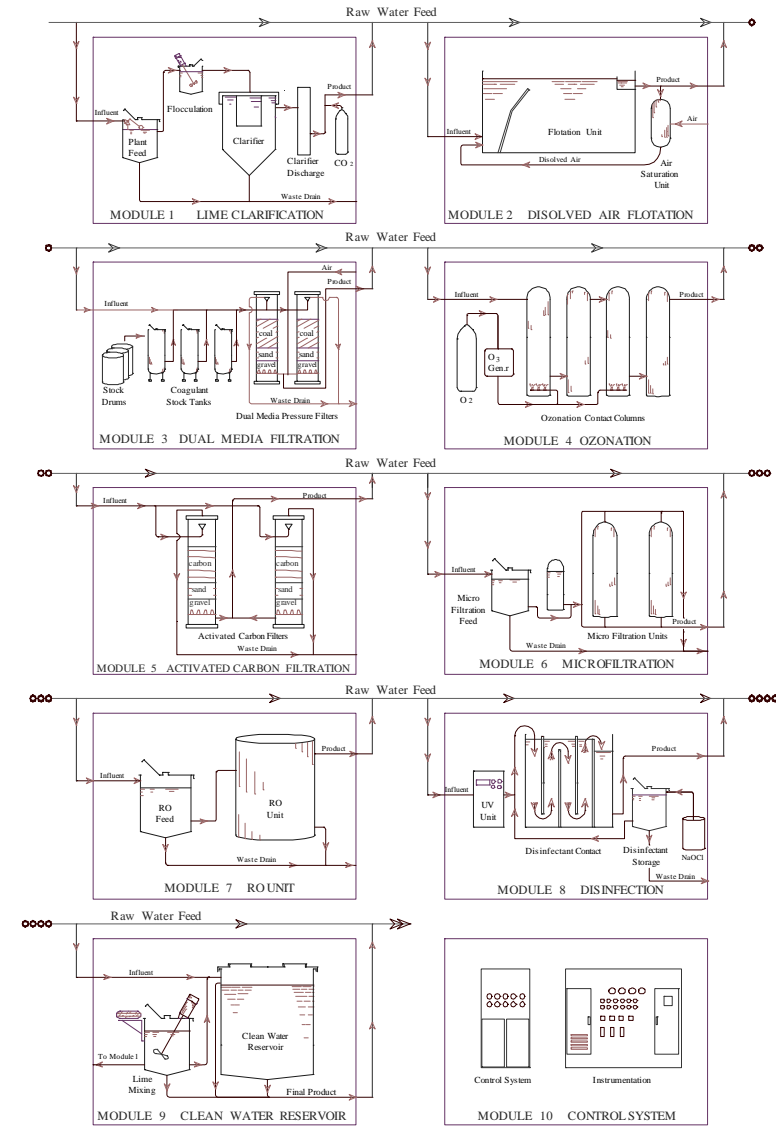


Figure 2 Diagrammatic layout of Advanced Water Recycling Demonstration Plant

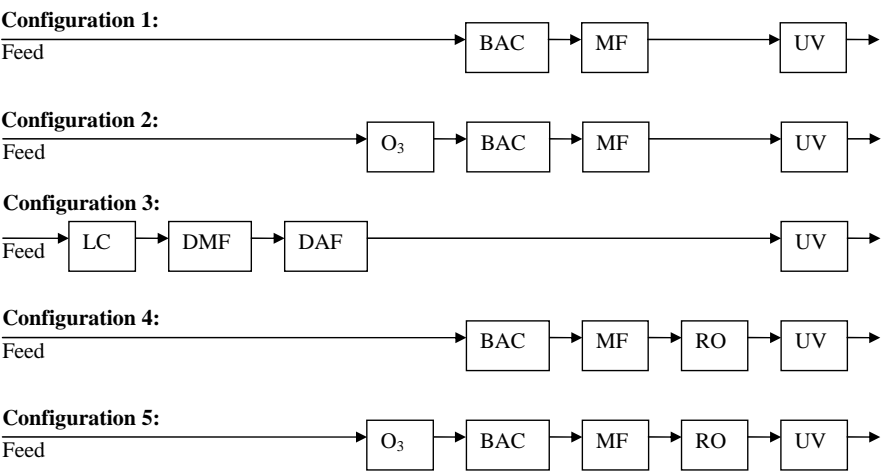


Figure 3 Plant configurations investigated in this study

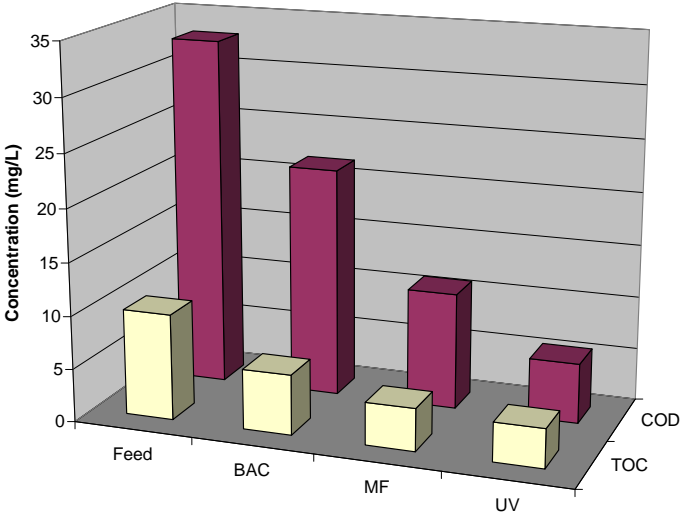


Figure 4: Bulk organic parameters removal with plant Configuration 1

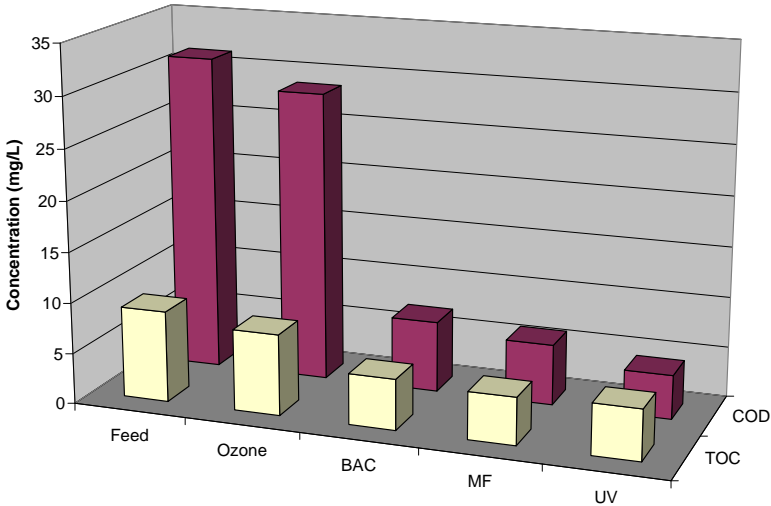


Figure 5: Bulk organic parameters removal with plant Configuration 2

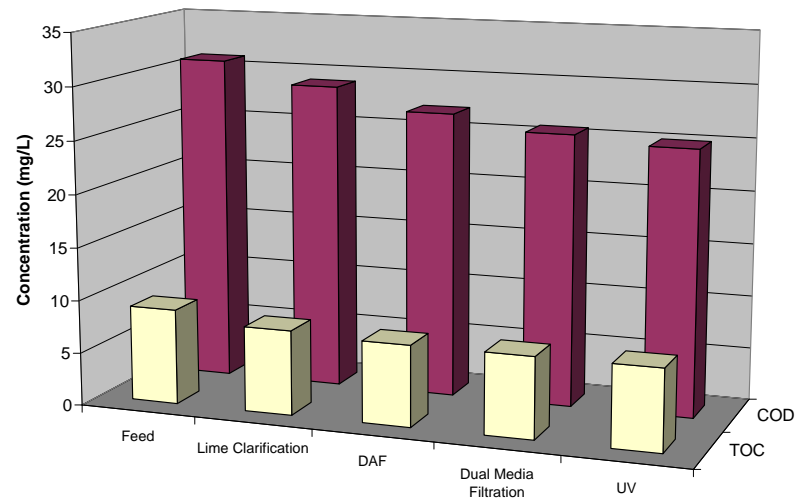


Figure 6: Bulk organic parameters removal with plant Configuration 3

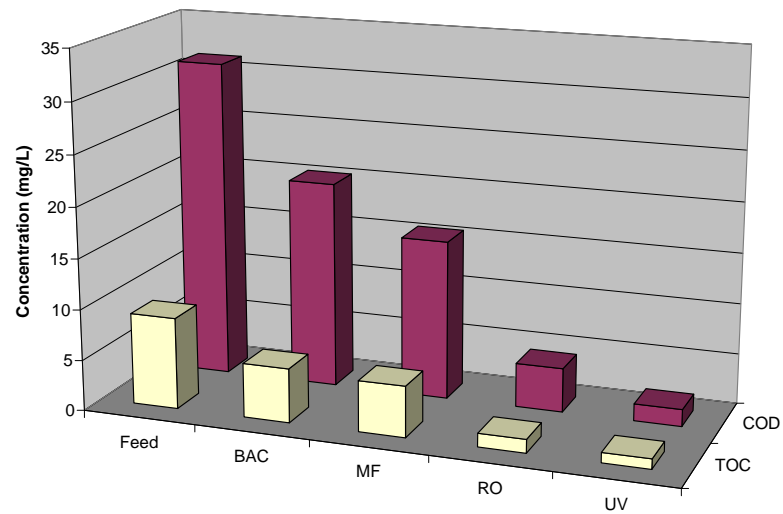


Figure 7: Bulk organic parameters removal with plant Configuration 4

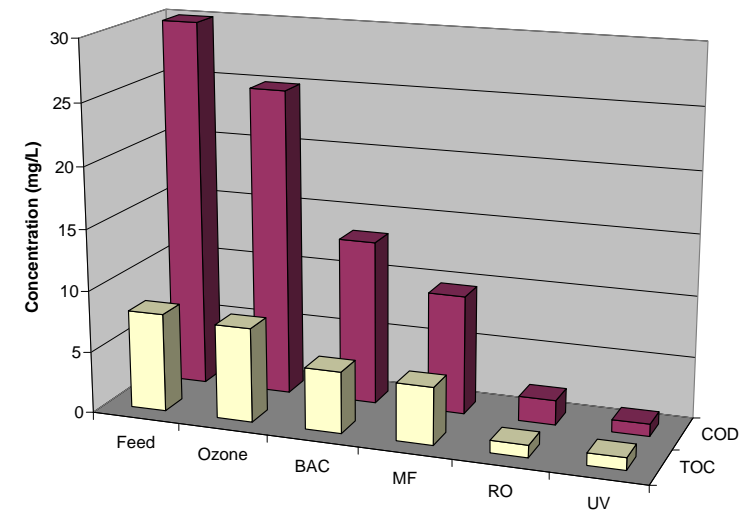


Figure 8: Bulk organic parameters removal with plant Configuration 5

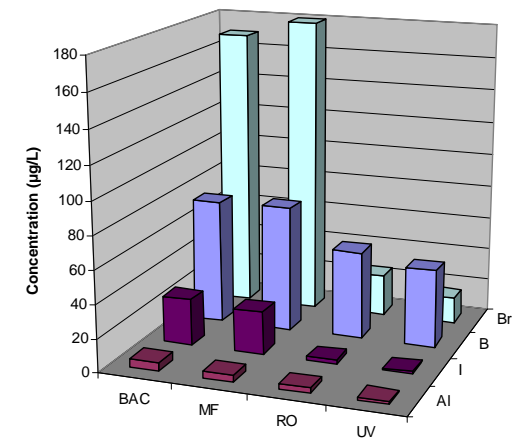


Figure 9: Inorganic parameters with plant Configuration 4